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10/621,399	07/18/2003	Koji Yonehara	21581-00298-US1	2449
30678 7590 09/19/2007 CONNOLLY BOVE LODGE & HUTZ LLP 1875 EYE STREET, N.W. SUITE 1100 WASHINGTON, DC 20036			EXAMINER OH, TAYLOR V	
			ART UNIT 1625	PAPER NUMBER
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**Please find below and/or attached an Office communication concerning this application or proceeding.**

The time period for reply, if any, is set in the attached communication.

## Office Action Summary

Application No.

10/621,399

Applicant(s)

YONEHARA ET AL.

Examiner

Taylor Victor Oh

Art Unit

1625

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

### Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

### Status

- 1) ☒ Responsive to communication(s) filed on 29 June 2007.
- 2a) ☐ This action is **FINAL**. 2b) ☒ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

### Disposition of Claims

- 4) ☒ Claim(s) 1,3-9,11-15 and 21 is/are pending in the application.
- 4a) Of the above claim(s) \_\_\_\_\_ is/are withdrawn from consideration.
- 5) ☐ Claim(s) \_\_\_\_\_ is/are allowed.
- 6) ☒ Claim(s) 1,3-9,11-15 and 21 is/are rejected.
- 7) ☐ Claim(s) \_\_\_\_\_ is/are objected to.
- 8) ☐ Claim(s) \_\_\_\_\_ are subject to restriction and/or election requirement.

### Application Papers

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on \_\_\_\_\_ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.  
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).  
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

### Priority under 35 U.S.C. § 119

- 12) ☒ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☒ All b) ☐ Some \* c) ☐ None of:
1. ☒ Certified copies of the priority documents have been received.
2. ☐ Certified copies of the priority documents have been received in Application No. \_\_\_\_\_.
3. ☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

\* See the attached detailed Office action for a list of the certified copies not received.

### Attachment(s)

- 1) ☐ Notice of References Cited (PTO-892)
- 2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
- 3) ☐ Information Disclosure Statement(s) (PTO/SB/08)  
Paper No(s)/Mail Date \_\_\_\_\_
- 4) ☐ Interview Summary (PTO-413)  
Paper No(s)/Mail Date. \_\_\_\_\_
- 5) ☐ Notice of Informal Patent Application
- 6) ☐ Other: \_\_\_\_\_

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Applicant's arguments with respect to claims 1, 3-9, and 11-15, and 21 are considered but are moot in view of the new ground(s) of rejection.

**The Status of Claims :**

Claims 1, 3-9, and 11-15, and 21 are pending.

Claims 1, 3-9, and 11-15 and 21 are rejected.

**DETAILED ACTION**

1. Claims 1, 3-9, and 11-15, and 21 are under consideration in this Office Action.

Priority

2. It is noted that the foreign priority document , Japan 2002-209987 is in the file.

Drawings

3. None.

***Claim Rejections - 35 USC § 112***

The following is a quotation of the first paragraph of 35 U.S.C. 112:

The specification shall contain a written description of the invention, and of the manner and process of making and using it, in such full, clear, concise, and exact terms as to enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to make and use the same and shall set forth the best mode contemplated by the inventor of carrying out his invention.

Claims 11-15 are rejected under 35 U.S.C. 112, first paragraph, as failing to comply with the enablement requirement. The claim(s) contains subject matter which was not described in the specification in such a way as to enable one skilled in the art to which it pertains, or with which it is most nearly connected, to make and/or use the invention. "The factors to be considered [in making an enablement rejection] have been summarized as the quantity of experimentation necessary, the amount of direction or guidance presented, the presence or absence of working examples, the nature of the invention, the state of the prior art, the relative skill of those in that art, the predictability or unpredictability of the art, and the breadth of the claims"

Claims are drawn to the generic method of preventing any tungsten species leaching out from any solid catalyst form. The meaning of preventing any tungsten species leaching out from any solid catalyst form is the absolute zero % of tungsten species leaching out during the process. However, applicants are not enabled for preventing a tungsten species leaching during the process of carrying out liquid-phase oxidation reaction using the catalyst containing tungsten species. There are very few selected examples such as  $W/Zn-SnO_2$  ,  $W/Zn-SnO_2$  ,

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$\text{Fe}_2\text{SiW}_{10}\text{O}_{38}/\text{Zn-SnO}_2$  from tables 5, 6, and 7 with zero % leaching in the specification, whereas the other examples in the tables 1-3 are far from showing the true prevention of the tungsten species leaching during the process. Thus, there is no evidence of record that would guide the skilled artisan to identify which catalyst containing the tungsten species can be surely prevented from leaching out. The Examiner suggests deletion of the word "preventing" and the phrase "reducing the risk". Therefore, an appropriate correction is required.

The following is a quotation of the second paragraph of 35 U.S.C. 112:

The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

Claim 21 is rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

In claim 21, the phrase "containing 2 to 15" is recited. The term "containing" is vague and indefinite because the term "comprising" would mean that there are other components besides tungsten atom; there is uncertainty as to what other compounds are present in the product. Furthermore, it is well-settled that the term "containing" do not exclude the presence of other ingredients than the one recited. Ex parte Muench, 79 USPQ 92 (PTO Bd. App. 1948). Therefore, an appropriate correction is required.

***Claim Rejections - 35 USC § 102***

The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless –

b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

1. Claims 1,4-5, 7-9, and 21 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Neumann et al (WO 98/54165).

Neumann et al discloses the followings (see pages 6-7):

The process described in this invention relates to the use of transition metal substituted polyoxometalates (TMSP) to catalyze the epoxidation of alkenes with molecular oxygen according to the following equation (5).



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Polyoxometalates are oligomeric oxides of defined structure based on addenda of tungsten, molybdenum, niobium or vanadium or a combination thereof. More specifically, transition metal substituted polyoxometalates are compounds of the general formula  $X_x(TM)_yM_mO_z^{q-}$  where the heteroatom, X, if present ( $x \geq 0$ ) may be main group or transition metals, the addenda atoms, M, are molybdenum, tungsten, niobium or vanadium or a combination thereof, and TM is one or several different transition metals. The specific class of transition metal substituted polyoxometalates,  $[WZnTM_2(XW_9O_{19})_2]^{q-}$ , used in the process described by the present invention are characterized as a dimer of a truncated Keggin structure having a "belt" of W, Zn and other transition metal (TM) cations "sandwiched" between the two  $B-XW_9O_{34}$  trivacant Keggin fragments the structure of which is shown in appended Figure 1. The transition metal cations are assumed to be positioned at terminal positions and are hexacoordinate with at least one labile ligand such as water. The TM atom can be any transition metal of the first, second or third row. More preferably the TM atom is a noble metal selected from the group consisting of ruthenium, rhodium, palladium, osmium, iridium and platinum. Most preferably the TM atom is ruthenium. The heteroatom atom, X, of the trivacant Keggin fragments can be any heteroatom known for Keggin compounds as is well-known in the art. For example, X can be a nonmetal such as phosphorous, silicon, germanium, boron, or arsenic. Alternatively X can be a metal such as zinc, cobalt, iron etc. The preferred transition metal substituted polyoxometalate for this process is  $[WZnRu_2(ZnW_9O_{19})_2]^{11-}$ . No transition metal substituted polyoxometalates or those of the general structure described and shown above have ever been used as catalysts for the epoxidation of alkenes with molecular oxygen. The counter cation of the above transition metal substituted polyoxometalates may be any cation including for example alkali metals, alkaline earth cations, transition metal cations or organic cations such as quaternary ammonium salts.

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**Example 1**

A 5 ml solution of degassed 1,2-dichloroethane containing 100  $\mu\text{mol}$   $\text{Q}_{11}\text{WZnRu}_2(\text{ZnW}_9\text{O}_{19})_2$  in which Q is tricaprylmethyl ammonium was kept under 1 atm molecular oxygen at 90 °C for nine hours in a closed vessel. The oxygen solution was cooled to room temperature and 11.2 mg cyclooctene was added.

After 2 hours the solution was analyzed by GLC. The analysis showed a 67.5 % conversion to cyclooctene oxide.

(see page 8, example 1). Furthermore, it shows that the third element can be Fe and Zn (see pages 6-7). This is identical with the claims.

2. Claims 1,3-7, 9, and 21 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Brown et al (WO 94/21583).

Brown et al discloses the followings (see from page 3 ,line 18 to page 4, line 24):



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According to the present invention, there is provided a process for the selective oxidation of alcohols wherein a substrate alcohol is contacted with hydrogen peroxide in a reaction medium in the presence of catalyst, characterised in that:

- i. the reaction medium comprises a solvent that does not comprise a carboxyl group, and
- ii. the catalyst comprises a heteropolyacid which is supported on an insoluble particulate support.

In the process according to the present invention, the catalyst contains a non-metallic heteroatom which may be selected from Group IV including silicon and germanium or Group V including phosphorus. Preferably, the catalyst contains phosphorus as the non-metallic heteroatom.

Tungsten or molybdenum, preferably tungsten, may constitute the entire metallic component of the heteropolyacid. Two transition metals, such as tungsten and molybdenum, may be incorporated therein. Many heteropolyacids for employment in the present invention process can be represented by the empirical formula  $Q_{3/n}PW_wMo_{12-w}O_{40}$  when they are brought into contact with the support in which  $w$  represents 0 or an integer of at least 1 and often at least 6. Most preferably,  $w$  represents 12.  $Q$  represents hydrogen or other counterion, and  $n$  is its basicity in the general formula. It is believed that the supported catalyst retains its empirical ratio of tungsten to phosphorous and molybdenum, but that the interaction of the catalyst with the surface of the support may result in the catalyst becoming bonded chemically to the support, thereby modifying both the catalyst itself and the support surface. Such treatments may also encourage a redistribution of the metal between species of different nuclearity.

Other heteropolyacids contemplated for use in the present invention include those containing at least one first series transition metal, including specifically iron, manganese, cobalt and nickel, for example in heteropolyacids of the formula  $Q(7-v)/nPW_{11}MxO_{36+v}$  in which  $Mx$  represents the other transition metal,  $v$  is its oxidation state and  $Q$  is the counterion of basicity  $n$  as before.

The support for the heteropolyacid catalyst is often selected from solids of Group IIa, IIb, IIIb, IVa and IVb elements and from organic basic ion exchange resins. Within the class of Group IIa compounds, it is often convenient to select as support from magnesium compounds, including in particular, magnesium oxide and magnesium silicate. Within the class of group IIb compounds, it can be convenient to select from zinc compounds as support, including specifically zinc oxide. Within the class of Group IVa compounds, it is often convenient to select from titanium or zirconium compounds, including specifically titanium oxide and zirconium phosphate. From within the class of inorganic Group IVb compounds, it is often desirable to select as supports from tin compounds, many of which are readily available, or from germanium compounds. Specific examples include tin oxide.

During calcination of the supported catalyst material which has been obtained by impregnation of an inorganic support with the heteropolyacid, it is believed that formation of a bond between the catalyst and the support is promoted, which can assist in controlling the leaching of catalyst into the reaction mixture. However, as a result of such interaction and bond formation, the catalyst species may be altered to some extent by such calcination, so that the value of such a post impregnation calcination tends to vary depending upon the support employed. For some supports, including alumina in particular, it is advantageous to calcine at a temperature of at least 300°C and usually not higher than about 600°C. In a number of instances, a particularly suitable temperature for post-impregnation calcination of for example alumina is at least about 400°C and especially from about 450 to about 550°C. Other supports for which post impregnation calcination is an appropriate activity include magnesium silicate and zirconium phosphate. (s

ee page 6 ,lines 22-35). This is identical with the claims.

3. Claims 1,4-5, 7-9, and 21 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Watanabe et al (J. of Molecular Catalysis A : Chem.. 145 (1999), p. 281-289) discloses the followings (see an abstract page 281):

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The epoxidation of alkenes with hydrogen peroxide or molecular oxygen, in the presence of aldehyde catalyzed by heteropolyoxometalate (HPOM) salts and layered double hydroxides (LDHs) containing HPOM anions as pillars, has been investigated. In the case of oxidation with hydrogen peroxide, a lacunary Keggin-ion,  $\text{SiW}_{11}\text{O}_{39}^{8-}$ , was more active than  $\text{SiW}_{12}\text{O}_{40}^{4-}$ . For this reaction system using hydrogen peroxide, intercalated  $\text{SiW}_{11}\text{O}_{39}^{8-}$  showed higher stability in structure and activity than free  $\text{K}_x\text{SiW}_{11}\text{O}_{39}$ . For the oxidation using molecular oxygen and aldehyde,  $\text{SiW}_{12}\text{O}_{40}^{4-}$  showed higher activity than  $\text{SiW}_{11}\text{O}_{39}^{8-}$ . It was confirmed that  $\text{SiW}_{12}\text{O}_{40}^{4-}$  was intercalated between the brucite-type layers of the LDHs without change in structure. Ni-containing LDHs without any interstitial polyoxometalate showed high activity for the oxidation of cyclohexene with the combination of  $\text{O}_2$  and aldehyde. © 1999 Elsevier Science B.V. All rights reserved.

### 2.3.1. Oxidation of alkenes with $\text{H}_2\text{O}_2$

Oxidation reactions of alkenes were carried out in a 100-ml flask containing catalyst (1 mmol of polymetal), alkene (100 ml) and 10 wt.%  $\text{H}_2\text{O}_2$  (100 mmol) in tributyl phosphate (TBP), at 343 K for 3 h. A non-aqueous  $\text{H}_2\text{O}_2$ -TBP solution was prepared by adding 30%  $\text{H}_2\text{O}_2$  aqueous solution to TBP, followed by evaporation of water under reduced pressure at 323 K [16].

Table 1  
Chemical compositions and methods of synthesis

Sample	Chemical composition (molar ratio) $\text{M}^{2+}:\text{Al}^{3+}:\text{metal}^a$
$\text{Zn}_2\text{Al-SiW}_{12}\text{O}_{40}$	2.0:1.0:0.44
$\text{Zn}_3\text{Al-SiW}_{12}\text{O}_{40}$	3.0:1.0:0.78
$\text{Zn}_4\text{Al-SiW}_{12}\text{O}_{40}$	3.3:1.0:0.12
$\text{Zn}_3\text{Al-SiW}_{11}\text{O}_{39}$	2.9:1.0:1.4
$\text{Zn}_2\text{Al-SiW}_9\text{O}_{14}$	2.0:1.0:1.2
$\text{Mg}_2\text{Al-SiW}_{12}\text{O}_{40}$	2.3:1.0:1.2
$\text{Ni}_2\text{Al-SiW}_{12}\text{O}_{40}$	1.3:1.0:1.6
$\text{Ni}_2\text{Al-TA}^b$	2.2:1.0:0

<sup>a</sup> Polymetal in HPOM.

<sup>b</sup> TA = terephthalate dianion.

This is identical with the claims.

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4. Claims 1,4-5, 7-9 , and 21 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Brown et al (WO 93/00338) discloses the followings (see an abstract page):

Alkenes can be epoxidised with hydrogen peroxide using a homogeneous heavy metal catalyst, but discharge of spent reaction mixtures releases the heavy metal in the environment. The problem can be ameliorated by selecting a heterogeneous catalyst system comprising a tungsten-containing heteropolyacid supported on selected Group IIa, IIb, IIb, IVa or IVb inorganic supports or on a strong basic resin, which catalyst has either been calcined after impregnation or in the impregnation stage an alcoholic solution of the heteropolyacid is employed and by employing a nitrilo or oxygenated polar solvent reaction medium. A number of preferred heteropolyacids satisfy the empirical formula  $M_{3/n}PW_wMO_{12-w}O_{40}$  in which w represents an integer of at least 1, M represents a counterion and n its basicity. Preferred supports include activated alumina, calcined at 400 to 600° C and cross-linked quaternary ammonium-substituted polystyrene resins. The most preferred catalysts are made by impregnating an inorganic support with a methanol solution of the heteropolyacid to a desired loading of active material on the support and subsequently calcining the loaded support at 400 to 600° C. Preferred reaction media include acetonitrile and tertiary butanol.

#### Examples 1 to 8 and Comparisons C9 to C14

In these Examples and Comparisons, the selected catalyst (0.5g of as made catalyst/support), substrate (cyclohexene, 2g except in Ex 3, Ex4, Ex7 and C11, C15 which each employed 8g) and solvent (40ml) were charged into a three-necked glass reaction vessel equipped with a water-cooled condenser and the stirred mixture was heated to reflux temperature, which was about 80°C, except in Example 2, where it was about 60°C. Aqueous hydrogen peroxide solution (35% w/w except in Ex3 which used 70% w/w) was introduced slowly into the stirred refluxing reaction mixture over a period of 45 minutes to 1 hour. The mixture was refluxed for a further 5 hours. The reaction mixture was then analysed by gas liquid chromatography to determine how much substrate had been consumed during the reaction, including evaporation losses,

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Table 1

Catalyst Ref.	Impregnation Species	Support	Prep Method	Calcine Temp °C
I	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	γ-Al <sub>2</sub> O <sub>3</sub>	A	500
II	H <sub>3</sub> PW <sub>6</sub> Mo <sub>11</sub> O <sub>40</sub>	"	A	500
III	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	neutral Al <sub>2</sub> O <sub>3</sub>	A	500
IV	"	Mg silicate	A	500
V	(CP) <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	γ-alumina	B	500
VI	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	γ-Al <sub>2</sub> O <sub>3</sub>	A	600
VII	"	"	A	400
VIII	"	"	A	200
IX	"	"	A	-
X	"	"	A	500
XI	"	"	B	500
XII	"	ZrPO <sub>4</sub>	A	500
XIII	"	SnO <sub>2</sub>	A	500
XIV	"	Al(OH) <sub>3</sub>	A	500
XVI	"	TiO <sub>2</sub>	A	500
XVII	H <sub>3</sub> PW <sub>6</sub> Mo <sub>6</sub> O <sub>40</sub>	α-alumina	A	500
R1	H <sub>3</sub> PMo <sub>12</sub> O <sub>40</sub>	SiO <sub>2</sub>	A	500
R2	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	acid-Al <sub>2</sub> O <sub>3</sub>	A	500
R3	"	basic-Al <sub>2</sub> O <sub>3</sub>	A	500
R4	"	α-alumina	A	500
R5	"	H-mordenite	A	500
R6	"	not supported		


This is identical with the claims.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Taylor Victor Oh whose telephone number is 571-272-0689. The examiner can normally be reached on 8:30-5:00.

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If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Janet Andres can be reached on 571-272-0867. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free). If you would like assistance from a USPTO Customer Service Representative or access to the automated information system, call 800-786-9199 (IN USA OR CANADA) or 571-272-1000.

  
Taylor Victor Oh, MSD, LAC  
Primary Examiner  
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9/17/07